

JCN07 Rec'd PCT/PTO 04 DEC 2001

FORM PTO-1390 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			2895-0131P
INTERNATIONAL APPLICATION NO. PCT/JP00/03582			U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <b>09/980586</b>
INTERNATIONAL FILING DATE June 2, 2000		PRIORITY DATE CLAIMED June 4, 1999	

**TITLE OF INVENTION**  
NON-AQUEOUS ELECTROLYTE SOLUTION, ELECTRODE, AND CAPACITOR CONSTRUCTED THEREOF

**APPLICANT(S) FOR DO/EO/US**

MITA, Sakoto; TAKAMUKU, Yoshinori

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau. WO 00/75941
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is transmitted herewith.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4)
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 20. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98, Form PTO-1449(s), and International Search Report (PCT/ISA/210) with 9 cited document(s).
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:
  - 1.) PCT/IB/304
  - 2.) PCT/IB/308
  - 3.) One (1) sheet of Formal Drawings

U.S. APPLICATION NO (if known, see 37 CFR 1.5) <b>09/980586</b>	INTERNATIONAL APPLICATION NO PCT/JP00/03582	ATTORNEY'S DOCKET NUMBER 2895-0131P
--	--	--

21. <input checked="" type="checkbox"/> The following fees are submitted:				<b>CALCULATIONS</b>		<b>PTO USE ONLY</b>	
<b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO. ....				\$1,040.00			
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO. ....				\$890.00			
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. ....				\$740.00			
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) .....				\$710.00			
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) .....				\$100.00			
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>					\$	890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).					\$	0	
<b>CLAIMS</b>	<b>NUMBER FILED</b>	<b>NUMBER EXTRA</b>	<b>RATE</b>				
Total Claims	20 - 20 =	0	X \$18.00	\$	0		
Independent Claims	1 - 3 =	0	X \$84.00	\$	0		
MULTIPLE DEPENDENT CLAIM(S) (if applicable) Yes				+ \$280.00	\$	280.00	
<b>TOTAL OF ABOVE CALCULATIONS =</b>					\$	1170.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.					\$	0	
<b>SUBTOTAL =</b>					\$	1170.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).					\$	0	
<b>TOTAL NATIONAL FEE =</b>					\$	1170.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +					\$	40.00	
<b>TOTAL FEES ENCLOSED =</b>					\$	1210.00	
					Amount to be:		
					refunded	\$	
					charged	\$	

☒ A check in the amount of \$ **1210.00** to cover the above fees is enclosed.


☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_ to cover the above fees.  
A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 02-2448.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
1.37(a) or (b)) must be filed and granted to restore the application to pending status.

Send all correspondence to:  
Richard Stewart, Kolasch & Birch, LLP or Customer No. 2292  
P.O. Box 747  
Falls Church, VA 22040-0747  
Phone (703) 205-8000

Date: December 4, 2001

By   
Marc S. Weiner, #32,181

09/980586

JC13 Rec'd PCT/PTO 0 4 DEC 2001

PATENT  
2895-0131P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: MITA, Sakoto et al.  
Int'l. Appl. No.: PCT/JP00/03582  
Appl. No.: New Group:  
Filed: December 4, 2001 Examiner:  
For: NON-AQUEOUS ELECTROLYTIC SOLUTION,  
ELECTRODE, AND CAPACITOR  
CONSTRUCTED THEREOF

PRELIMINARY AMENDMENT

**BOX PATENT APPLICATION**

Assistant Commissioner for Patents  
Washington, DC 20231

December 4, 2001

Sir:

The following Preliminary Amendments and Remarks are respectfully submitted in connection with the above-identified application.

AMENDMENTS

IN THE SPECIFICATION:

Please amend the specification as follows:

Before line 1, insert --This application is the national phase under 35 U.S.C. § 371 of PCT International Application No. PCT/JP00/03582 which has an International filing date of June 2, 2000, which designated the United States of America.--

IN THE CLAIMS:

Please amend the claims as follows:

8. (Amended) A non-aqueous electrolytic solution for a capacitor as claimed in claim 1, wherein said solvent for the electrolyte comprises a mixed solvent prepared by admixing the compound of the formula (2) with a cyclic carbonic ester and/or phosphoric ester.

9. (Amended) A non-aqueous electrolytic solution for a capacitor as claimed in claim 1, wherein said solvent for the electrolyte comprises the compound of the formula (2) by 0.01-50% by weight.

10. (Amended) A non-aqueous electrolytic solution for a capacitor as claimed in claim 1, wherein said capacitor is an electric double layer capacitor.

11. (Amended) A capacitor which comprises a non-aqueous electrolytic solution for a capacitor as claimed in claim 1.

12. (Amended) An electric double layer capacitor which comprises a non-aqueous electrolytic solution for a capacitor as claimed in claim 1.

13. (Amended) An electrode for a capacitor which is pre-treated with an electrolytic solution as claimed in claim 1.

REMARKS

The specification has been amended to provide a cross-reference to the previously filed International Application.

The claims have been amended to delete improper multiple dependencies and to place the application into better form for examination.


Entry of the above amendments is earnestly solicited. An early and favorable first action on the merits is earnestly solicited.

Attached hereto is a marked-up version of the changes made to the application by this Amendment.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By   
Marc S. Weiner, #32,181

MSW/cqc  
2895-0131P

P.O. Box 747  
Falls Church, VA 22040-0747  
(703) 205-8000

Attachment: VERSION WITH MARKINGS TO SHOW CHANGES MADE

VERSION WITH MARKINGS TO SHOW CHANGES MADE

The claims have been amended as follows:

8. (Amended) A non-aqueous electrolytic solution for a capacitor as claimed in [any claim of claims 1-7]claim 1, wherein said solvent for the electrolyte comprises a mixed solvent prepared by admixing the compound of the formula (2) with a cyclic carbonic ester and/or phosphoric ester.

9. (Amended) A non-aqueous electrolytic solution for a capacitor as claimed in [any claims of claim 1-8]claim 1, wherein said solvent for the electrolyte comprises the compound of the formula (2) by 0.01-50% by weight.

10. (Amended) A non-aqueous electrolytic solution for a capacitor as claimed in [any claim of claims 1-9]claim 1, wherein said capacitor is an electric double layer capacitor.

11. (Amended) A capacitor which comprises a non-aqueous electrolytic solution for a capacitor as claimed in [any claim of claims 1-9]claim 1.

12. (Amended) An electric double layer capacitor which comprises a non-aqueous electrolytic solution for a capacitor as claimed in [any claim of claims 1-9]claim 1.

13. (Amended) An electrode for a capacitor which is pre-treated with an electrolytic solution as claimed in [any claim of claims 1-9]claim 1.

(Rev. 11/13/01)



1/p<sup>st</sup> 1**Specification****Non-aqueous Electrolytic Solution, Electrode, and Capacitor  
Constructed Thereof**

5

**Technical Field of the Invention:**

The present invention relates to a silicone compound-  
containing non-aqueous electrolytic solution for a capacitor,  
an electrode and a capacitor constructed thereof, and more  
10 particularly a non-aqueous electrolytic solution for a  
capacitor having high energy density and excellent withstand  
voltage and charging/discharging cycle properties, an electrode  
for a capacitor, and a capacitor constructed thereof.

15 **Background of the Invention:**

An electric double layer capacitor, being a low-power direct  
current power source having an intermediate capacity ranked  
between a battery and an ordinary capacitor, is widely used as  
a backup power source for IC's and memories and a supplement  
20 or alternative to secondary batteries. Such new ideas have been  
conceived recently as to utilize electric double layer  
capacitors in newly introduced portable electronic devices such  
as VTR with a built-in camera, cellular phones and laptop  
computers. It is due to said trend that there is a demand  
25 mounting for development of electric double layer capacitors  
having higher energy densities.

The electric double layer capacitor, different from the

battery which takes advantage of chemical reactions to generate electric energy, takes advantage of large capacities of electric double layers created in the interface between electrodes and the electrolytic solution, so as to serve as a power source by inputting/outputting electric charges generated in the double layers similarly to the process of charging/discharging of electricity in the case of the battery.

Such electric double layer capacitor is constructed of two leaves of electrode, a separator, and generally a corrosion resistant electrolytic solution. Namely, such capacitor is of such construction as is constructed of two leaves of electrode formed from a material having large surface areas such as activated carbon and a binder such as fluoroplastic which are disposed face-to-face with a porous separator being interposed in between with spaces between the electrodes and the porous separator and an interior of the porous separator, respectively, being filled up with an electrolytic solution.

Included in the category of such electrolytic solution are an aqueous solution-based electrolytic solution and an organic solvent-based electrolytic solution (non-aqueous electrolytic solution). The aqueous electrolytic solution having a withstand voltage of as low as about 1.2 V is not suitable for the electric double layer capacitor for which a high energy density is required. On the contrary, the organic solvent-based electrolytic solution (non-aqueous electrolytic solution) affords a possibility of producing a capacitor having a high energy density by virtue of its higher withstand voltage

compared with the aqueous electrolytic solution.

The non-aqueous electrolytic solutions have been conventionally manufactured generally by mixing and dissolving an electrolyte, such as a tetrafluoroboric acid compound and tetraethyl ammonium compound, in a non-aqueous solvent such as cyclic carbonic ester having a high dielectric constant. Nevertheless, inasmuch as the cyclic carbonic ester solvent at an elevated temperature undergoes decarboxylation decomposition reaction which is normally accompanied by generation of a large volume of gases, it is said to shorten service life of the capacitor. Furthermore, such electrolytic solution not necessarily possesses a sufficient withstand voltage, and hence further improvement in charging/discharging cycle properties has been wanted.

#### **Description of the Invention:**

Accordingly, it is an object of the present invention to provide a non-aqueous electrolytic solution for a capacitor which releases little amount of gases from a solvent for an electrolyte, possesses high withstand voltage and excellent charging/discharging cycle properties, and excels in its safety. It is another object of the present invention to provide an electrode for a capacitor and a capacitor in which said non-aqueous electrolytic solution is employed.

The present invention relates to a non-aqueous electrolytic solution for a capacitor comprising an electrolyte and a solvent

for the electrolyte comprising a compound having therein a silicon-containing group represented by the following formula (1), particularly a non-aqueous electrolytic solution for an electric double layer capacitor.

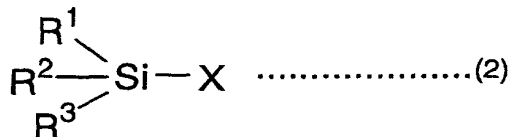


5

Wherein  $\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  are each a monovalent group selected from the group consisting of hydrogen atom, halogen atom, hydrocarbon group having 1 - 6 carbon atoms, and oxyhydrocarbon group having 1 - 6 carbon atoms, wherein a hydrocarbon portion of said hydrocarbon group or oxyhydrocarbon group may be of a chain structure or cyclic structure and may be saturated or unsaturated and part of hydrogen atoms in the hydrocarbon portion may be substituted with halogen atom, and  $\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  may be the same or different.

15

The compound having therein the silicon-containing group represented by the formula (1) is preferably a compound represented by the following formula (2).



20

Wherein X is, as is mentioned later, a group selected from the group consisting of oxazolidinone group, imidazole group, phosphate group, phenyl group, alkoxy group, halogen atom, unsaturated alkyl group, carbamate group, and carbonate group, etc.

Among the above-identified compounds, particularly preferred as the solvent for electrolyte are N-trialkylsilylimidazole, tris(trialkylsilyl) phosphate, 4-trialkylsilylmethyl-1,3-dioxolan-2-one, trialkylchlorosilane, tri-  
5 fluoroalkyltrialkylsilane, allyltrialkylsilane, diallyl-  
dialkylsilane, triallylalkylsilane, acetyltrialkylsilane, alkyl-N-trialkylsilyl carbamate, alkyl-N,N-bistrialkylsilyl carbamate, alkyltrialkylsilyl carbonate, and 3-trialkylsilyl-2-oxazolidinone.

10 It is preferable that said compound having therein the silicon-containing group is used in the form of a mixed solvent prepared with another solvent, such as cyclic carbonic ester and phosphoric ester, in such manner that it is admixed by 0.01-50 % by weight in the mixed solvent.

15 Additionally, the present invention relates to a capacitor containing said non-aqueous electrolytic solution, particularly an electric double layer capacitor.

Furthermore, the present invention relates to an electrode for a capacitor which is pre-treated with said non-aqueous  
20 electrolytic solution and a capacitor into which such electrode is incorporated.

#### Brief Description of the Drawings:

Figure 1 is a perspective view illustrating the construction  
25 of the roll-up type electric double layer capacitor element employed in Examples.

Figure 2 is a perspective view illustrating the mode of

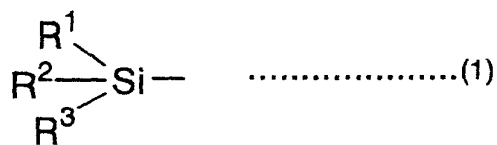
inserting the roll-up type electric double layer capacitor element employed in Examples into a metal container together with a closure member.

## 5 Detailed Description of the Preferred Embodiments:

Detailed explanations are set forth as follows about the construction of the non-aqueous electrolytic solution, the electrode, and the capacitor constructed thereof. The non-aqueous electrolytic solution comprises of the electrolyte and  
 10 the solvent for such electrolyte, and is suitable for the capacitor, particularly the electric double layer capacitor.

### Silicon Compounds

The non-aqueous electrolytic solution of the present  
 15 invention comprises a compound having therein a silicon-containing group as represented by the following formula (1) as the solvent for electrolyte.



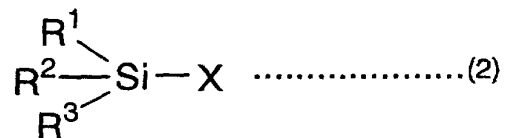
Wherein  $\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  are each a monovalent group selected  
 20 from the group consisting of hydrogen atom, halogen atom, hydrocarbon group having 1 - 6 carbon atoms such as alkyl group, and oxyhydrocarbon group having 1 - 6 carbon atoms such as alkoxy group, wherein a hydrocarbon portion of said hydrocarbon group or oxyhydrocarbon group may be of a chain structure or cyclic  
 25 structure like that of phenyl group, and may be saturated or

unsaturated and part of hydrogen atoms in the hydrocarbon portion may be substituted with halogen atom.

$R^1$ ,  $R^2$ , and  $R^3$  may be preferably a saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms, or more preferably an alkyl group having 1 - 3 carbon atoms. Part or all of hydrogen atoms in the alkyl group may be substituted with halogen atom, particularly fluorine atom. Said groups may be the same or different, but it is preferable that they are the same.

As specific examples of said groups there can be cited trimethylsilyl group, triethylsilyl group, tripropylsilyl group, trifluoromethylsilyl group and trimethoxysilyl group.

The compound having therein the silicon-containing group represented by the formula (1) is preferably a compound represented by the following formula (2).



Wherein X is optionally selected from among the formulae (3) - (14) which are cited as follows, and constitutes any compound identified as (a) - (1).

20

( a ) Oxazolidinone compounds:



In the formula (3), part of hydrogen atoms may be substituted

with saturated or unsaturated hydrocarbon group having 1- 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group or halogen atom. Wherein the hydrocarbon group may have its part or all hydrogen atoms substituted with halogen atoms.

- 5 As specific examples of compounds having therein said groups there can be cited 3-trimethylsilyl-2-oxazolidinone, 3-trimethylsilyl-4-trifluoromethyl-2-oxazolidinone, and 3-triethylsilyl-2-oxazolidinone.

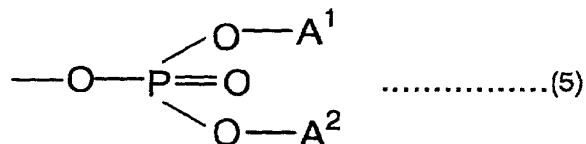
10 ( b ) Imidazole compounds:



- In the formula (4), part of hydrogen atoms may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group or halogen atom.
- 15

As specific examples of compounds having therein said groups there can be cited N-trimethylsilylimidazole, N-trimethylsilyl-4-methylimidazole, and N-triethylsilylimidazole.

20 ( c ) Phosphate compounds:



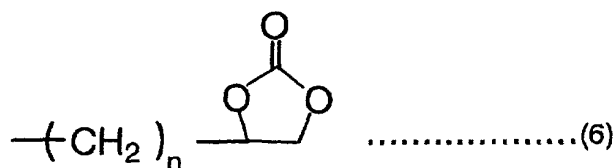
In the formula (5), A<sup>1</sup> and A<sup>2</sup> are each a saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms, or the



silicon-containing group represented by the formula (1), which may be the same or different.

As specific examples of compounds having therein said groups there can be cited tris(trimethylsilyl) phosphate, tris-  
 5 (triethylsilyl) phosphate, trimethylsilyldimethyl phosphate, and trimethylsilyldiallyl phosphate.

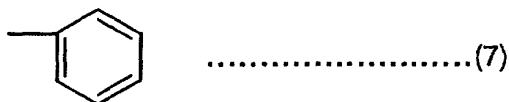
( d ) Cyclic carbonate compounds:



10 In the formula (6), part of hydrogen atoms which are bonded to carbon atoms constituting the cyclic structure may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group, halogen atom, or the silicon-containing group  
 15 represented by the formula (1), wherein "n" is a number of 0, 1, or 2.

As specific examples of compounds having therein said groups there can be cited 4-trimethylsilyl-1,3-dioxolan-2-one, 4-trimethylsilyl-5-vinyl-1,3-dioxolan-2-one, and 4-trimethyl-  
 20 silylmethyl-1,3-dioxolan-2-one.

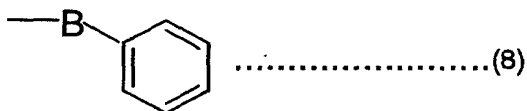
( e ) Phenyl compounds (1):



In the formula (7), part of hydrogen atoms which are bonded to carbon atoms constituting the benzene ring may be substituted with a saturated or unsaturated hydrocarbon group having 1 -  
 5 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group, halogen atom, or the silicon-containing group represented by the formula (1).

As specific examples of compounds having therein said groups there can be cited phenyltrimethylsilane, phenyltriethylsilane,  
 10 and phenyltrimethoxysilane.

( f ) Phenyl compounds (2):



In the formula (8), B is any bivalent group selected from  
 15 the group consisting of - NH -, - N(CH<sub>3</sub>) -, - CH<sub>2</sub> -, - CO -, - S -, and - SO<sub>2</sub> -, wherein part of hydrogen atoms which are bonded to carbon atoms constituting the benzene ring may be substituted with a saturated or unsaturated hydrocarbon group having 1-  
 4 carbon atoms, alkoxy group, amino group, cyano group, nitro  
 20 group, halogen atom, or the silicon-containing group represented by the formula (1).

As specific examples of compounds having therein said groups there can be cited phenylthiotrimethylsilane and phenyl-

thiotriethylsilane.

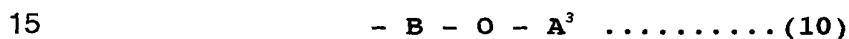
( g ) Ether compounds (1):



5        In the formula (9),  $A^3$  is a monovalent saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms, or the silicon-containing group represented by the formula (1), wherein said hydrocarbon group may be of a chain structure or cyclic structure.

10       As specific examples of compounds having therein said groups there can be cited methoxytrimethylsilane, hexamethyldisiloxane, and pentamethyldisiloxane.

( h ) Ether compounds (2):

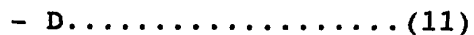


In the formula (10),  $A^3$  and B are the same as the groups shown in the aforementioned formulae (8) and (9), respectively.

As specific examples of compounds having therein said groups there can be cited methoxymethyltrimethylsilane.

20

( i ) Silane compounds:

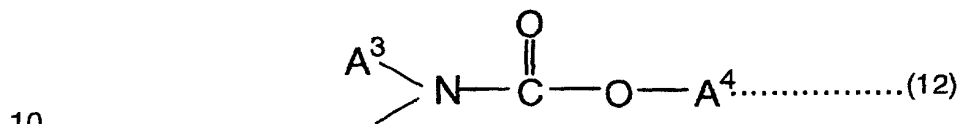


25       In the formula (11), D is any group selected from the group consisting of halogen atom, hydrogen atom, a saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms, acetyl group and the silicon-containing group represented by the formula (1). Wherein said hydrocarbon group may be of a chain

structure or cyclic structure and part of hydrogen atoms in the group may be substituted with halogen atom or alkoxy group.

As specific examples of compounds having therein said groups there can be cited trimethylchlorosilane, butyldiphenylchlorosilane, trifluoromethyl-trimethylsilane, acetyl-trimethylsilane, 3-trimethylsilylcyclopentene, allyl-trimethylsilane, and vinyltrimethylsilane.

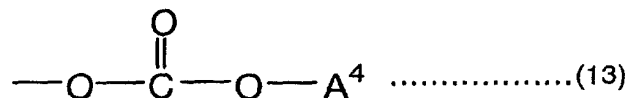
( j ) Carbamate compounds:



In the formula (12), A<sup>3</sup> is the same as the group shown in the aforementioned formula (9) and A<sup>4</sup> is a saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms.

As specific examples of compounds having therein said groups there can be cited methyl-N-trimethylsilyl carbamate, methyl-N,N-bistrimethylsilyl carbamate, ethyl-N-trimethylsilyl carbamate, methyl-N-triethylsilyl carbamate, and vinyl-N-trimethylsilyl carbamate.

( k ) Chain carbonate compounds:



In the formula (13), A<sup>4</sup> is the same as the group shown in the aforementioned formula (12).

As specific examples of compounds having therein said groups there can be cited methyltrimethylsilyl carbonate, allyltrimethylsilyl carbonate, and ethyl-trimethylsilyl carbonate.

5 ( 1 ) Other compounds:

- B - D .....(14)

In the formula (14), B and D are the same as the groups shown in the aforementioned formulae (8) and (11), respectively.

As specific examples of compounds having therein said groups  
10 there can be cited hexamethyldisilazane.

Among the aforementioned compounds having therein a silicon-containing group, the compound of (b), (c), (d), (i), (j) and (k) are preferable.

15

Out of the compounds, preferred for the present invention is any compound selected from the group consisting of N-trialkylsilylimidazole, tris(trialkylsilyl) phosphate, 4-trialkylsilylmethyl-1,3-dioxolan-2-one, trialkylchlorosilane,  
20 trifluoroalkyltrialkylsilane, allyltrialkylsilane, diallyl-dialkylsilane, triallylalkylsilane, acetyltrialkylsilane, alkyl-N-trialkylsilyl carbamate, alkyl-N,N-bis-trialkylsilyl carbamate, and alkyltrialkylsilyl carbonate.

As preferred specific examples among the compounds there  
25 can be cited N-trimethylsilylimidazole, tris(trimethylsilyl) phosphate, allyltrimethylsilane, 4-trimethylsilylmethyl-1,3-dioxolan-2-one, methyl-N-trimethylsilyl carbamate,

methyl-N,N-bis(trimethylsilyl) carbamate and methyl-trimethylsilyl carbonate.

In the present invention 3-trialkylsilyl-2-oxazolidinone such as 3-trimethylsilyl-2-oxazolidinone is preferably used,  
5 too.

A single kind of silicon compound may be used individually or two or more kinds may be used in combination.

The silicon compound shown in the formula (2) is chemically  
10 stable and highly acid-proof, will be hardly oxidized when allowed to stand in the air, and is less likely to react with water or such highly reactive substance as metallic lithium under normal storage conditions. Furthermore, such silicon compound possesses such properties that it is physically stable  
15 and resistive to thermal decomposition, exhibits low flammability and is hardly susceptible to electrochemical oxidation and reduction. Besides, those silicon compounds have relatively high flash point, and hence, their safety against fires is excellent.

20

#### Solvent for Electrolyte

According to the present invention said silicon compound is used singularly or in the form of a mixed solvent prepared in combination with another solvent as the solvent for  
25 electrolyte. In cases where it is used in the form of the mixed solvent it is preferable that the silicon compound shown in the aforementioned formula (2) is admixed with another solvent by

0.01- 50 % by weight, or preferably 0.1 - 30 % by weight, in the solvent. Since electrochemically and physically stable solvent for electrolyte is prepared, the obtained solvent gives improved flame resistance so long as the mixing ratio is adjusted within said ranges.

As such other solvent that can be used in said mixed solvent, any compound may be optionally selected from among the below-exemplified cyclic carbonic esters, chain carbonic esters, cyclic esters, chain esters, cyclic ethers, chain ethers, phosphoric esters, and sulfur-containing compounds.

(1) Cyclic carbonic esters:

Ethylene carbonate (syn.: 1,3-dioxolan-2-one), propylene carbonate (syn.: 4-methyl-1,3-dioxolan-2-one), butylene carbonate (syn.: 4,5-dimethyl-1,3-dioxolan-2-one), and vinylene carbonate, etc.

Cyclic carbonic esters may be also those which have alkyl groups wherein hydrogen atoms are substituted with halogen atoms such as monofluoromethylethylene carbonate, difluoromethylethylene carbonate, and trifluoromethylethylene carbonate.

(2) Chain carbonic esters:

Dimethyl carbonate, methylethyl carbonate, diethyl carbonate, methylpropyl carbonate, and methylisopropyl carbonate, etc.

(3) Cyclic esters:

$\gamma$ -butyrolactone,  $\gamma$ -valerolactone, 3-methyl- $\gamma$ -butyro-

lactone, and 2-methyl- $\gamma$ -butyrolactone, etc.

(4) Chain esters:

Methyl formate, ethyl formate, methyl acetate, ethyl acetate, propyl acetate, methyl propionate, methyl butyrate, and methyl valerate, etc.

(5) Cyclic ethers:

1,4-Dioxane, 1,3-dioxolane, tetrahydrofuran, 2-methyl-tetrahydrofuran, 3-methyl-1,3-dioxolane, and 2-methyl-1,3-dioxolane, etc.

10 (6) Chain ethers:

1,2-Dimethoxyethane, 1,2-diethoxyethane, diethyl ether, dimethyl ether, methylethyl ether, and dipropyl ether, etc.

(7) Phosphoric esters:

Trimethyl phosphate, triethyl phosphate, and triallyl phosphate, etc.

(8) Sulfur-containing compounds:

Sulfolane, etc.

It is preferable to use cyclic carbonic ester and/or phosphoric ester among the solvents with a view to furthermore improving charging/discharging cycle properties and low flammability of the solvent. Moreover, those solvents may be used singularly or in combination of two or more kinds.

25

Electrolyte

Ammonium salts, phosphonium salts and the like can be used as the electrolyte contained in the non-aqueous electrolytic



solution of the present invention. They may be used either singularly or in combination of two or more kinds. As specific examples of such electrolytes there can be cited the following compounds.

5

(1) Ammonium salts:

Tetrabutylammonium tetrafluoroborate  $((C_4H_9)_4 NBF_4)$ ,  
 tetraethylammonium tetrafluoroborate  $((C_2H_5)_4 NBF_4)$ ,  
 triethyl-monomethylammonium tetrafluoroborate  $((C_2H_5)_3 (CH_3) NBF_4)$ ,  
 10 tetrabutylammonium hexafluorophosphate  $((C_4H_9)_4 NPF_6)$ ,  
 tetraethylammonium hexafluorophosphate  $((C_2H_5)_4 NPF_6)$ , and  
 triethyl-monomethylammonium hexafluorophosphate  $((C_2H_5)_3 (CH_3) NPF_6)$ , etc.

(2) Phosphonium salts:

15 Tetrabutylphosphonium tetrafluoroborate  $((C_4H_9)_4 PBF_4)$ ,  
 tetraethylphosphonium tetrafluoroborate  $((C_2H_5)_4 PBF_4)$ ,  
 tetrabutylphosphonium hexafluorophosphate  $((C_4H_9)_4 PPF_6)$ , and  
 tetraethylphosphonium hexafluorophosphate  $((C_2H_5)_4 PPF_6)$ , etc.

(3) Electrolytes described in International Publication No. WO  
 20 95/15572, etc.:

Among the compounds preferred ones are tetrabutylammonium tetrafluoroborate  $((C_4H_9)_4 NBF_4)$ , tetraethylammonium tetrafluoroborate  $((C_2H_5)_4 NBF_4)$ , and triethyl-monomethylammonium tetrafluoroborate  $((C_2H_5)_3 (CH_3) NBF_4)$ . It is preferable that the  
 25 electrolyte is contained in the eletrolytic solution by a concentration of generally 0.1 - 3 mole/liter, or preferably 0.3 - 1.5 mole/liter.

### Electrodes and Capacitor

The non-aqueous electrolytic solution of the present invention is used for capacitors at large including the aluminum electrolytic capacitor, and particularly it is used suitably for the electric double layer capacitor.

The construction of the electric double layer capacitor is such that it is composed of two leaves of electrodes, a separator, and generally a corrosion resistant electrolytic solution. Specifically, two leaves of electrodes are disposed face-to-face with a separator being interposed in between; and the spaces between the electrode and the porous separator and the interior of the porous separator are filled with the electrolytic solution.

The electrode is constructed of a material having a large surface area like activated carbon and a binder such as fluoroplastic. The kinds of such activated carbon as is preferably used as the material for the electrode are generally phenolic-based, pitch-based, polyacrylonitrile-based, palm shell-derived or coke-based in either the filament form or the powder form. Normally, activated carbon having a specific surface area of 1000 m<sup>2</sup>/g or more is preferred. Such activated carbon is used after it has been activated by the steam activation method or the alkali activation method.

As the binder there are used such materials as carboxymethyl cellulose, poly(vinylidene fluoride), poly(vinylpyrrolidone), polyimide, poly(vinyl alcohol), and polyacrylic acid. The

material for the electrode is bound by a binder and rendered into an intended shape to be obtained as a fabricated electrode.

The electrode is also put in use after it has been pre-treated with said non-aqueous electrolytic solution. The  
5 pre-treatment may be carried out by spraying the non-aqueous electrolytic solution onto the electrode or by immersing the electrode in the non-aqueous electrolytic solution. It is by this pre-treatment that the efficiency of wetting may be increased between the electrode and the non-aqueous  
10 electrolytic solution and the stability of the electrode surface may be enhanced, and then the capacity of the capacitor may be that much more increased. It is preferable that the thus pre-treated electrode is used at least on either side of the anode or the cathode, while such electrode may be used in various  
15 capacitors at large besides the electric double layer capacitor.

The electrode is often used in a combination of said material for the electrode (the active substance) and a collector body. As such collector body, for example, aluminum foil is preferably used. As for the separator, on the other hand, preferably used  
20 are such materials as porous film made from polyethylene, polypropylene, and the like, kraft paper, manila paper, and glass filter.

The non-aqueous electrolytic solution of the present invention may also be usable for a metal surface treatment of  
25 the collector body, and then a metal corrosion of the collector body is restrained by the treatment.

An explanation about the construction of, for example, a

roll-up type electric double layer capacitor element is furnished as follows. Namely, it is of such construction that the separator is interposed between the polarizable anode to which an anode-side conducting wire is connected and the polarizable cathode to which a cathode-side conducting wire is connected, which in a three-layer stock are rolled up together. Rubber-made closure members are prepared for threading the anode-side conducting wire and the cathode-side conducting wire therethrough, respectively. After the capacitor element has been immersed in a driving electrolytic solution, it is inserted into an aluminum cylindrical container provided with a bottom. By means of carrying out horizontal drawing and curling jobs at the opening part of the metal container, the closure members are tightly sealed to the opening part of the metal container.

### Examples

The present invention will be illustrated in details with reference to the following examples; however, the scope of the invention is not limited by the examples.

Explanations are furnished as follows in reference to Figure 1 and Figure 2, respectively, about the method of fabricating the roll-up type electric double layer capacitor used in Examples and Comparative Example.

The capacitor element (1) was fabricated as illustrated in Figure 1 by rolling up together into a cylindrical shape the polarizable anode (3), the polarizable cathode (5) and the

separator (6) being interposed in between the two. Said polarizable electrodes were each produced by adding acetylene black and carboxymethyl cellulose to an activated carbon powder, dispersing the mixed powder in water to reduce to a slurry, coating the slurry onto an aluminum foil whose surface had been roughened in advance, and then drying the coated foil.

The capacitor element (1) was impregnated with the electrolytic solution prepared in accordance with the procedure specified in each Example, and was inserted into the cylindrical aluminum container (8) provided with bottom as illustrated in Figure 2. In the meantime, the anode-side conducting wire (2) was connected to the polarizable anode (3), and the cathode-side conducting wire (4) was connected to the polarizable cathode (5). Each conducting wire was put through the rubber-made closure member (7), and then the closure member (7) was attached to the opening part of the cylindrical container (8). The opening part was tightly sealed with the closure member (7) by horizontal drawing and curling jobs. Thus was fabricated a roll-up type electric double layer capacitor (9). The thus fabricated capacitor had a diameter of 18 mm and a height of 40 mm.

#### Example 1

25 ml of an electrolytic solution having an electrolyte concentration of 0.5 mole/liter was prepared by dissolving 2.71 g (0.0125 mole) of tetraethylammonium tetrafluoroborate ( $(C_2H_5)_4NBF_4$ ) in a mixed solvent prepared by admixing propylene carbonate

with N-trimethylsilylimidazole at a ratio of 95 : 5 by weight. The obtained electrolytic solution was tested with respect to a leakage current. Test results are shown in Table 1.

The leakage current was determined in accordance with the following procedure. Said electrolytic solution was put into a tripolar withstand voltage tester cell in which activated carbon electrodes were installed as an active electrode and an opposite electrode and an Ag/Ag<sup>+</sup> electrode was used as a reference electrode. Then, 2.0 V voltage was impressed on it, using a potentiogalvanostat. When a flow of electricity had become constant, an amperage was measured and recorded as the leakage current value.

Next, an electric double layer capacitor having a 2.3V and 50F rating was fabricated using said electrolytic solution. Thereupon, 2.5 V was impressed at an ambient temperature of 70°C, and a time-related dimensional change of the cylindrical container caused by gases built up in said atmosphere was measured as "a dimensional change of the product". The dimensional changes after the lapse of 500 hrs from the beginning of such elevated temperature load test are also shown in Table 1.

#### Example 2

The same procedure as in Example 1 was followed except that trimethyl phosphate was used in place of propylene carbonate as in Example 1 for determining the leakage current of the electrolytic solution and the dimensional change of the product

by the elevated temperature load test. Test results are shown in Table 1.

#### Example 3

- 5       The same procedure as in Example 1 was followed except that 3-trimethylsilyl-2-oxazolidinone was used in place of N-trimethylsilylimidazole as in Example 1 for determining the leakage current of the electrolytic solution and the dimensional change of the product by the elevated temperature load test.
- 10      Test results are shown in Table 1.

#### Example 4

- 15       The same procedure as in Example 1 was followed except that tris(trimethylsilyl) phosphate was used in place of N-trimethylsilylimidazole as in Example 1 for determining the leakage current of the electrolytic solution and the dimensional change of the product by the elevated temperature load test.
- Test results are shown in Table 1.

#### 20      Example 5

- The same procedure as in Example 4 was followed except that trimethyl phosphate was used in place of propylene carbonate as in Example 4 for determining the leakage current of the electrolytic solution and the dimensional change of the product
- 25      by the elevated temperature load test. Test results are shown in Table 1.

## Example 6

The same procedure as in Example 5 was followed except that a mixed solvent prepared by admixing trimethyl phosphate with tris(trimethylsilyl) phosphate at a ratio of 99 : 1 by weight was used for determining the leakage current of the electrolytic solution and the dimensional change of the product by the elevated temperature load test. Test results are shown in Table 1.

## 10 Example 7

The same procedure as in Example 2 was followed except that trimethylchlorosilane was used in place of N-trimethylsilylimidazole as in Example 2 for determining the leakage current of the electrolytic solution and the dimensional change of the product by the elevated temperature load test. Test results are shown in Table 1.

## Example 8

The same procedure as in Example 2 was followed except that allyltrimethylsilane was used in place of N-trimethylsilylimidazole as in Example 1 for determining the leakage current of the electrolytic solution and the dimensional change of the product by the elevated temperature load test. Test results are shown in Table 1.

25

## Example 9

The same procedure as in Example 2 was followed except that



methyl-N-trimethylsilylcarbamate was used in place of N-trimethylsilylimidazole as in Example 1 for determining the leakage current of the electrolytic solution and the dimensional change of the product by the elevated temperature load test.

5 Test results are shown in Table 1.

#### Comparative Example 1

10 An electrolytic solution was prepared in accordance with the same procedure as in Example 1 except that only propylene carbonate was used in place of a mixed solvent consisting of propylene carbonate and N-trimethylsilylimidazole as in Example 1. The thus prepared electrolytic solution was tested with respect to the leakage current of the electrolytic solution. Test results are shown in Table 1.

Table 1

	Kind of Solvent for Electrolyte (Ratio by weight)	Leakage Current ( $\mu$ A)	Dimensional Change of the Product (mm)
Example 1	Propylene carbonate vs. N-trimethylsilylimidazole = 95 : 5	180	1.5
Example 2	Trimethyl phosphate vs. N-trimethylsilylimidazole = 95 : 5	270	1.7
Example 3	Propylene carbonate vs. 3-trimethylsilyl-2- oxazolidinone = 95 : 5	270	1.6
Example 4	Propylene carbonate vs. Tris(trimethylsilyl)- phosphate = 95 : 5	90	1.4
Example 5	Trimethyl phosphate vs. Tris(trimethylsilyl)- phosphate = 95 : 5	140	1.5
Example 6	Trimethyl phosphate vs. Tris(trimethylsilyl)- phosphate = 99 : 1	250	1.6
Example 7	Trimethyl phosphate vs. Trimethylchlorosilane = 95 : 5	150	1.8
Example 8	Propylene carbonate vs. Allyltrimethylsilane = 95 : 5	80	1.2
Example 9	Propylene carbonate vs. Methy-N-trimethylsilyl- carbamate = 95 : 5	130	1.5
Comparative Example 1	Propylene carbonate = 100	280	2.1

From Table 1 it is known that the electrolytic solutions in Examples 1 - 9 exhibit less leakage current than that of Comparative Example 1, and hence superior with respect to the withstand voltage. Furthermore, they showed less dimensional changes of the product by the elevated temperature load test than that in case of Comparative Example 1.

#### **Possibility of the Industrial Utilization:**

The non-aqueous electrolytic solution of the present invention, by virtue of the silicon compound-containing solvent for electrolyte used therein, when it is utilized for the capacitor, proves itself to be the electrolytic solution which releases only little gases at an elevated temperature, gives high withstand voltage and exhibits excellent charging /discharging cycle properties. Additionally, said solvent for electrolyte has a higher flash point and it shows an excellent safety against fires.

The capacitor employing such non-aqueous electrolytic solution gives high withstand voltage, excellent charging/discharging cycle properties, and high energy density. Moreover, such capacitor has a long service life and the excellent safety against fires. On account of the above-mentioned advantages, such capacitor employing said non-aqueous electrolytic solution is suitable particularly for the electric double layer capacitor.

Furthermore, a capacity of the capacitor may be augmented due to increasing of the stability of the electrode surface by

treating the electrodes with such non-aqueous electrolytic solution in advance and thus may be obtained a capacitor having a higher efficiency.

What we claim is:

1. A non-aqueous electrolytic solution for a capacitor comprising an electrolyte and a solvent for the electrolyte comprising a compound having therein a silicon-containing group represented by the following formula (1),



wherein  $\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  are each a monovalent group selected from the group consisting of hydrogen atom, halogen atom, hydrocarbon group having 1 - 6 carbon atoms, and oxyhydrocarbon group having 1 - 6 carbon atoms, wherein a hydrocarbon portion of said hydrocarbon group or oxyhydrocarbon group may be of a chain structure or cyclic structure and may be saturated or unsaturated and part of hydrogen atoms in the hydrocarbon portion may be substituted with halogen atom, and

$\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  may be the same or different.

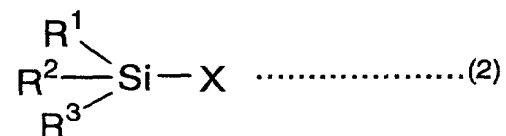
2. A non-aqueous electrolytic solution for a capacitor as claimed in claim 1, wherein  $\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  in the silicon-containing group of the formula (1) are each a saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms.

3. A non-aqueous electrolytic solution for a capacitor as claimed in claim 1, wherein  $\text{R}^1$ ,  $\text{R}^2$ , and  $\text{R}^3$  in the silicon-containing group of the formula (1) are each an alkyl group having

1 - 3 carbon atoms which are the same.

4. A non-aqueous electrolytic solution for a capacitor as claimed in claim 1, wherein  $R^1$ ,  $R^2$ , and  $R^3$  in the silicon-containing group of the formula (1) are selected from the group consisting of methyl group, ethyl group, and trifluoromethyl group.

5. A non-aqueous electrolytic solution for a capacitor as claimed in any claim of claims 1 - 3, wherein the compound having therein the silicon-containing group of the formula (1) is represented by the following formula (2),



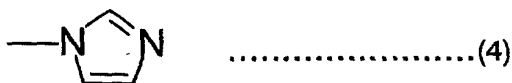
wherein X is any group selected from the group consisting of the groups represented by the following formulae (3) - (14), respectively,

(a)



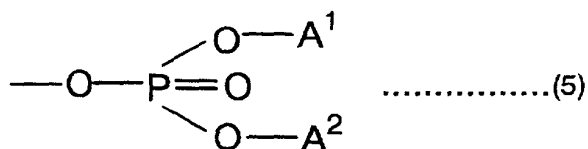
in the formula (3), part of hydrogen atoms may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group or halogen atom,

(b)



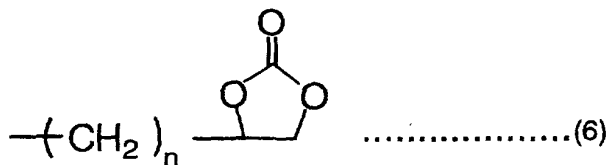
in the formula (4), part of hydrogen atoms may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group or halogen atom,

(c)



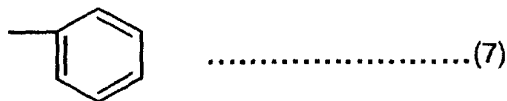
in the formula (5), A<sup>1</sup> and A<sup>2</sup> are each saturated or unsaturated hydrocarbon group having 1 - 6 carbon atoms or the silicon-containing group of the formula (1) which may be the same or different,

(d)



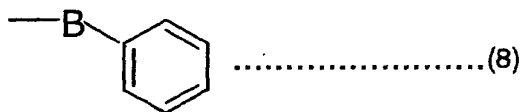
in the formula (6), part of hydrogen atoms which are bonded to carbon atoms constituting the cyclic structure may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group, halogen atom, or the silicon-containing group of the formula (1), wherein "n" is a number of 0, 1, or 2,

(e)



in the formula (7), part of hydrogen atoms which are bonded to carbon atoms constituting the benzene ring may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group, halogen atom, or the silicon-containing group of the formula (1),

(f)



in the formula (8), B is any bivalent group selected from the group consisting of - NH -, - N(CH<sub>3</sub>) -, - CH<sub>2</sub> -, - CO -, - S -, and - SO<sub>2</sub> -, wherein part of hydrogen atoms which are bonded to carbon atoms constituting the benzene ring may be substituted with a saturated or unsaturated hydrocarbon group having 1 - 4 carbon atoms, alkoxy group, amino group, cyano group, nitro group, halogen atom, or the silicon-containing group of the formula (1),

(g)



in the formula (9), A<sup>3</sup> is a saturated or unsaturated monovalent hydrocarbon group having 1 - 6 carbon atoms, or the silicon-containing group of the formula (1), wherein said hydrocarbon group may be of a chain structure or cyclic



structure,

(h)



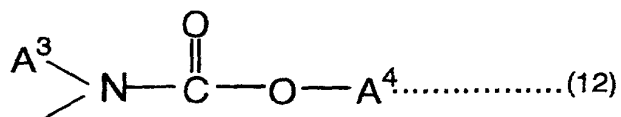
in the formula (10),  $A^3$  and B are the same as the groups shown  
5 in the formulae (8) and (9), respectively,

(i)



in the formula (11), D is any group selected from the group  
consisting of halogen atom, hydrogen atom, a saturated or  
10 unsaturated monovalent hydrocarbon group having 1 - 6 carbon  
atoms, acetyl group, and the silicon-containing group of the  
formula (1), wherein part of hydrogen atoms in said hydrocarbon  
group may be substituted with halogen atom or alkoxy group,

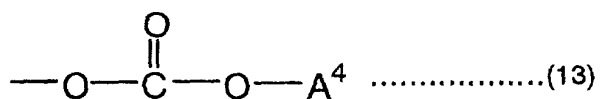
(j)



15

in the formula (12),  $A^3$  is the same as the group shown in the  
aforementioned formula (9) and  $A^4$  is a saturated or unsaturated  
hydrocarbon group having 1 - 6 carbon atoms,

(k)



20

in the formula (13),  $A^4$  is the same as the group shown in the  
formula (12),

and

(1)

- B - D..... (14)

in the formula (14), B and D are the same as the groups shown in the formulae (8) and (11), respectively.

5

6. A non-aqueous electrolytic solution for a capacitor as claimed in claim 5, wherein the compound of the formula (2) is any compound selected from the group consisting of N-trialkylsilylimidazole, tris(trialkylsilyl) phosphate, 4-trialkylsilylmethyl-1,3-dioxolan-2-one, trialkylchlorosilane, trifluoroalkyltrialkylsilane, allyltrialkylsilane, diallyl-dialkylsilane, triallylalkylsilane, acetyltrialkylsilane, alkyl-N-trialkylsilyl carbamate, alkyl-N,N-bis-trialkylsilyl carbamate, alkyltrialkylsilyl carbonate, and 3-trialkyl-silyl-2-oxazolidinone.

15

7. A non-aqueous electrolytic solution for a capacitor as claimed in claim 5, wherein the compound of the formula (2) is any compound selected from the group consisting of N-trimethylsilylimidazole, tris(trimethylsilyl) phosphate, allyltrimethylsilane, 4-trimethylsilylmethyl-1,3-dioxolan-2-one, methyl-N-trimethylsilyl carbamate, methyl-N,N-bis-trimethylsilyl carbamate, methyltrimethylsilyl carbonate, and 3-trimethylsilyl-2-oxazolidinone.

20

25

8. A non-aqueous electrolytic solution for a capacitor as claimed in any claim of claims 1 - 7, wherein said solvent for

the electrolyte comprises a mixed solvent prepared by admixing the compound of the formula (2) with a cyclic carbonic ester and/or phosphoric ester.

- 5 9. A non-aqueous electrolytic solution for a capacitor as claimed in any claim of claims 1 - 8, wherein said solvent for the electrolyte comprises the compound of the formula (2) by 0.01 - 50 % by weight.
- 10 10. A non-aqueous electrolytic solution for a capacitor as claimed in any claim of claims 1 - 9, wherein said capacitor is an electric double layer capacitor.
11. A capacitor which comprises a non-aqueous electrolytic  
15 solution for a capacitor as claimed in any claim of claims 1 - 9.
12. An electric double layer capacitor which comprises a non-aqueous electrolytic solution for a capacitor as claimed  
20 in any claim of claims 1 - 9.
13. An electrode for a capacitor which is pre-treated with an electrolytic solution as claimed in any claim of claims 1 - 9.
- 25 14. A capacitor wherein at least one electrode partly constituting such capacitor is an electrode as claimed in claim 13.

Fig. 1

1/1

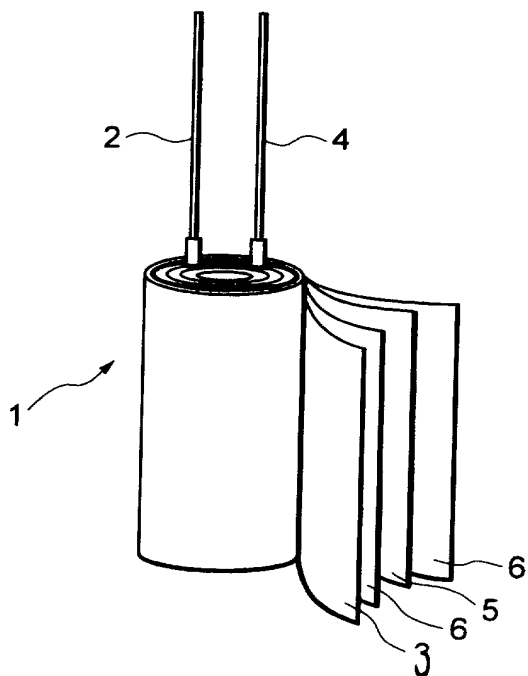
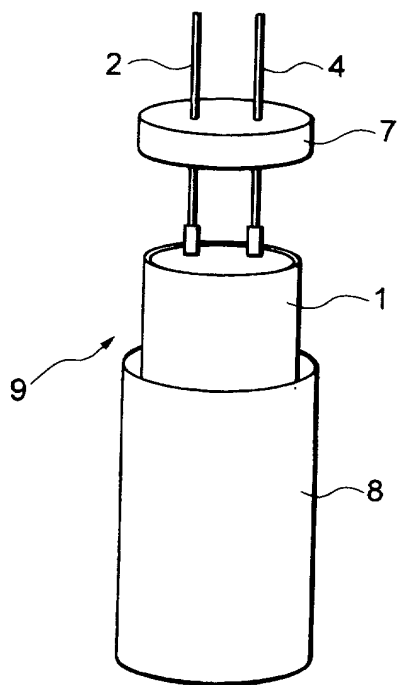


Fig. 2



# BIRCH, STEWART, KOLASCH & BIRCH, LLP

PLEASE NOTE:  
YOU MUST  
COMPLETE THE  
FOLLOWING:

## COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT AND DESIGN APPLICATIONS

ATTORNEY DOCKET NO.  
2895-0131P

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated next to my name; that I verily believe that I am the original, first and sole inventor ( if only one inventor is named below) or an original, first and joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Insert Title:

Non-aqueous Electrolytic Solution, Electrode, and Capacitor  
Constructed Thereof

Fill in Appropriate  
Information -  
For Use Without  
Specification  
Attached:

the specification of which is attached hereto. If not attached hereto,

the specification was filed on \_\_\_\_\_ as  
United States Application Number \_\_\_\_\_; and /or

the specification was filed on June 2, 2000 as PCT  
International Application Number PCT /JP00 /03582; and was  
amended under PCT Article 19 on \_\_\_\_\_ (if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I do not know and do not believe the same was ever known or used in the United States of America before my or our invention thereof, or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application, that the same was not in public use or on sale in the United States of America more than one year prior to this application, that the invention has not been patented or made the subject of an inventor's certificate issued before the date of this application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months (six months for designs) prior to this application, and that no application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application by me or my legal representatives or assigns, except as follows.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 (a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Insert Priority  
Information:  
(if appropriate)

Prior Foreign Application(s)

(Number)	(Country)	(Month/Day/Year Filed)	Priority Claimed
<u>157604/1999</u>	<u>Japan</u>	<u>06/04/1999</u>	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
____	____	____	<input type="checkbox"/> Yes <input type="checkbox"/> No
____	____	____	<input type="checkbox"/> Yes <input type="checkbox"/> No
____	____	____	<input type="checkbox"/> Yes <input type="checkbox"/> No
____	____	____	<input type="checkbox"/> Yes <input type="checkbox"/> No
____	____	____	<input type="checkbox"/> Yes <input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, §119(e) of any United States provisional application(s) listed below.

Insert Provisional  
Application(s):  
(if any)

(Application Number)	(Filing Date)
____	____
____	____

All Foreign Applications, if any, for any Patent or Inventor's Certificate Filed More Than 12 Months (6 Months for Designs) Prior To The Filing Date of This Application:

Insert Requested  
Information:  
(if appropriate)

Country	Application No	Date of Filing (Month/Day/Year)
____	____	____
____	____	____

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

Insert Prior U.S.  
Application(s):  
(if any)

(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)
____	____	____
____	____	____

I hereby appoint the following attorneys to prosecute this application and/or an international application based on this application and to transact all business in the Patent and Trademark Office connected therewith and in connection with the resulting patent based on instructions received from the entity who first sent the application papers to the attorneys identified below, unless the inventor(s) or assignee provides said attorneys with a written notice to the contrary:

15  
Terrell C. Birch (Reg. No. 19,382)  
Joseph A. Kolasch (Reg. No. 22,463)  
Bernard L. Sweeney (Reg. No. 24,448)  
Charles Gorenstein (Reg. No. 29,271)  
Leonard R. Svensson (Reg. No. 30,330)  
Andrew D. Meikle (Reg. No. 32,868)  
Joe McKinney Muncy (Reg. No. 32,334)  
C. Joseph Faraci (Reg. No. 32,350)

Raymond C. Stewart (Reg. No. 21,066)  
James M. Slattery (Reg. No. 28,380)  
Michael K. Mutter (Reg. No. 29,680)  
Gerald M. Murphy, Jr. (Reg. No. 28,977)  
Terry L. Clark (Reg. No. 32,644)  
Marc S. Weiner (Reg. No. 32,181)  
Donald J. Daley (Reg. No. 34,313)

Send Correspondence to:

**BIRCH, STEWART, KOLASCH & BIRCH, LLP**

**P.O. Box 747 • Falls Church, Virginia 22040-0747**

**Telephone: (703) 205-8000 • Facsimile: (703) 205-8050**

PLEASE NOTE:  
YOU MUST  
COMPLETE THE  
FOLLOWING:

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full Name of First or Sole  
Inventor:  
Insert Name of Inventor  
Insert Date This  
Document is Signed

Insert Residence  
Insert Citizenship

Insert Post Office  
Address

Full Name of Second  
Inventor, if any:  
see above

Full Name of Third  
Inventor, if any  
see above

Full Name of Fourth  
Inventor, if any  
see above

Full Name of Fifth  
Inventor, if any  
see above

GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Satoko	MITA	Satoko Mita	5. Nov. 2001
Residence (City, State & Country)		CITIZENSHIP	
Sodegaura-shi, Chiba, Japan		Japanese	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
c/o MITSUI CHEMICALS, INC. 580-32, Aza Taku 2-gou, Nagaura, Sodegaura-shi, Chiba 299-0265, Japan			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Yoshinori	TAKAMUKU	Yoshinori Takamuku	7. Nov. 2001
Residence (City, State & Country)		CITIZENSHIP	
Kyoto-shi, Kyoto, Japan		Japanese	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
Amenity Daigo 311, 21-2, Daigo Nakayama-cho, Fushimi-ku, Kyoto-shi, Kyoto 601-1373, Japan			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Residence (City, State & Country)		CITIZENSHIP	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Residence (City, State & Country)		CITIZENSHIP	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			
GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Residence (City, State & Country)		CITIZENSHIP	
POST OFFICE ADDRESS (Complete Street Address including City, State & Country)			

\* DATE OF SIGNATURE